

## **AMENDMENTS TO THE SPECIFICATION:**

On page 1, immediately following the title please insert headings as follows:

### **BACKGROUND OF THE INVENTION**

#### **Field of the Invention**

The paragraph beginning on page 1, line 4 has been changed as follows:

This invention ~~generally~~ relates generally to improved methods of fabricating molecular electronic devices, in particular organic electronic devices such as organic light emitting diodes (OLEDs) by droplet deposition techniques such as ink jet printing. The invention also relates to molecular device substrates fabricated by and/or use in such methods.

On page 1, line 9 please insert a heading as follows:

#### **Related Technology**

The paragraphs beginning on page 1, line 10 have been changed as follows:

Organic light emitting diodes (OLEDs) are a particularly advantageous form of electro-optic display. They are bright, ~~colourful~~ colorful, fast-switching, provide a wide viewing angle and are easy and cheap to fabricate on a variety of substrates. Organic (which here includes organometallic) LEDs may be fabricated using either polymers or small molecules in a range of ~~colours~~ colors (or in ~~multi-coloured~~ multi-colored displays), depending upon the materials used. A typical OLED device comprises two layers of organic material, one of which is a layer of light emitting material such as a light emitting polymer

(LEP), oligomer or a light emitting low molecular weight material, and the other of which is a layer of a hole transporting material such as a polythiophene derivative or a polyaniline derivative.

Organic LEDs may be deposited on a substrate in a matrix of pixels to form a single or ~~multi-colour~~ multi-color pixellated display. A ~~multicoloured~~ multi-colored display may be constructed using groups of red, green, and blue emitting pixels. So-called active matrix displays have a memory element, typically a storage capacitor and a transistor, associated with each pixel whilst passive matrix displays have no such memory element and instead are repetitively scanned to give the impression of a steady image.

The paragraph beginning on page 2, line 23 has been changed as follows:

A cathode layer 110 is then applied by, say, physical ~~vapour~~ vapor deposition. A cathode layer typically comprises a low work function metal such as calcium or barium covered with a thicker, capping layer of ~~aluminium~~ aluminum and optionally including an additional layer immediately adjacent the electroluminescent layer, such as a layer of lithium fluoride, for improved electron energy level matching. Mutual electrical isolation of cathode lines may achieved through the use of cathode separators (element 302 of Figure 3b). Typically a number of displays are fabricated on a single substrate and at the end of the fabrication process the substrate is scribed, and the displays separated before an encapsulating can is attached to each to inhibit oxidation and moisture ingress.

The paragraph beginning on page 4, line 5 has been changed as follows:

Figure 2 shows a view from above (that is, not through the substrate) of a portion of a ~~three-colour~~ three-color active matrix pixellated OLED display 200 after deposition of one of the active ~~colour~~ color layers. The figure shows an array of banks 112 and wells 114 defining pixels of the display.

The paragraphs beginning on page 4, 10 have been changed as follows:

Figure 3a shows a view from above of a substrate 300 for inkjet printing a passive matrix OLED display. Figure 3b shows a cross-section through the substrate of ~~figure~~ Figure 3a along line Y-Y'.

Referring to ~~figures~~ Figures 3a and 3b, the substrate is provided with a plurality of cathode undercut separators 302 to separate adjacent cathode lines (which will be deposited in regions 304). A plurality of wells 308 is defined by banks 310, constructed around the perimeter of each well 308 and leaving an anode layer 306 exposed at the base of the well. The edges or faces of the banks are tapered onto the surface of the substrate as shown, heretofore at an angle of between 10 and 40 degrees. The banks present a hydrophobic surface in order that they are not wetted by the solution of deposited organic material and thus assist in containing the deposited material within a well. This is achieved by treatment of a bank material such as polyimide with an O<sub>2</sub>/CF<sub>4</sub> plasma as disclosed in EP 0989778. Alternatively, the plasma treatment step may be avoided by use of a fluorinated material such as a fluorinated polyimide as disclosed in WO 03/083960.

The paragraph beginning on page 6, line 24 has been changed as follows:

Figure 4a shows a simplified cross section 400 through a well 308 filled with dissolved material 402, and ~~figure~~ Figure 4b shows the same well after the material has dried to form a solid film 404. In this example the bank angle is approximately  $15^\circ$  and the bank height is approximately  $1.5\mu\text{ m}$ . As can be seen a well is generally filled until it is brimming over. The solution 402 has a contact angle  $\theta_c$  with the plasma treated bank material of typically between  $30^\circ$  and  $40^\circ$  for example around  $35^\circ$ ; this is the angle the surface of the dissolved material 402 makes with the (bank) material it contacts, for example angle 402a in ~~figure~~ Figure 4a. As the solvent evaporates the solution becomes more concentrated and the surface of the solution moves down the tapering face of a bank towards the substrate; pinning of the drying edge can occur at a point between the initially landed wet edge and the foot of the bank (base of the well) on the substrate. The result, shown in ~~figure~~ Figure 4b, is that the film of dry material 404 can be very thin, for example of the order of 10nm or less, in a region 404a where it meets the face of a bank. In practice drying is complicated by other effects such as the coffee ring – effect. With this effect because the thickness of solution is less at the edge of a drop than in the centre, as the edge dries the concentration of dissolved material there increases. Because the edge tends to be pinned solution then flows from the centre of the drop towards the edge by capillary effect. This effect can result in dissolved material tending to be deposited in a ring rather than uniformly. The physics of the interactions of a drying solution with a surface are extremely complicated and a complete theory still awaits development.

The paragraph beginning on page 8, line 6 has been changed as follows:

Filling a well or pixel of a similar size to a drop presents little problem as when the drop lands it spreads out and fills the well. This is illustrated in ~~figure~~ Figure 5a which shows a well 500 for a long thin pixel of a type which is typically used in a RGB (red green blue) display. In the example of ~~figure~~ Figure 5a the pixel has a width of  $50\mu\text{m}$  and a length of  $150\mu\text{m}$  with  $20\mu\text{m}$  wide banks (giving a  $70\mu\text{m}$  pixel pitch and a  $210\mu\text{m}$  full ~~colour~~ color pitch). Such a well can be filled by three  $50\mu\text{m}$  droplets 502a, b, c as shown. Referring now to ~~figure~~ Figure 5b this shows a well 510 for a pixel which is approximately four times larger than each dimension giving a pixel width of approximately  $200\mu\text{m}$ , more suitable for applications such as a ~~colour~~ color television. As can be seen from the figure, many droplets 512 are needed to fill such a pixel. In practice, these tend to coalesce to form a larger droplet 514 which tends not to properly fill corners of the pixel (although Figures 5a and 5b and idealised and, in practice, the corners are not generally as sharp as they are shown). One way around this problem is to sufficiently over fill the well that the dissolved material well is pushed into the corners. This can be achieved by using a large number of dilute droplets and a high barrier around the well. Techniques for depositing large volumes of liquid are described in WO03/065474, which describes the use of very high barriers (for examples at page 8 lines 8 to 20) to allow the wells to hold a large volume of liquid without the liquid overflowing to adjacent wells. However such structures cannot easily be formed by photolithography and instead a plastic substrate is embossed or injection moulded. It is also desirable to be able to fill a well using fewer (higher concentration) droplets as this enables, *inter alia* faster printing.

On page 9, line 3 please add a heading as follows:

#### GENERAL DESCRIPTION OF THE INVENTION

The paragraph beginning on 9, line 4 has been changed as follows:

According to a first aspect of the ~~present~~ invention there is therefore provided a method of fabricating a molecular electronic device, the method comprising: fabricating a substrate having a plurality of banks defining wells for the deposition of molecular material; and depositing into said wells a composition comprising a molecular electronic material dissolved in a solvent, using a droplet deposition technique, to fabricate said device; wherein a said bank has a face, defining an edge of said well, at an angle to a base of the well of greater than a contact angle of said composition with said bank face; and wherein a height of a said bank above a said base of a said well is less than  $2\mu\text{m}$ , and more preferably less than  $1.5\mu\text{m}$ .

The paragraph beginning on page 11, line 13 has been changed as follows:

In a further aspect of the ~~present~~ invention there is therefore provided a method of fabricating a molecular electronic device, the method comprising: fabricating a substrate having a plurality of banks defining wells for the deposition of molecular material; and depositing into said wells a composition comprising a molecular electronic material dissolved in a solvent, using a droplet deposition technique, to fabricate said device; wherein a said bank has a face, defining an edge of said well, at an angle to a base of the well of at least  $40^\circ$ ; and wherein a height of a said bank above a said base of a said well is less than  $2\mu\text{m}$ , and

more preferably less than 1.5 $\mu$ m. Preferably, the angle is at least 50°. The angle may be up to 90° or, in some embodiments, greater than 90°.

On page 14, line 3 please insert a heading as follows:

#### BRIEF DESCRIPTION OF THE DRAWINGS

The paragraph beginning on page 14, line 10 has been changed as follows:"

Figure 2 shows a view from above of a portion of a three ~~colour~~ color pixelated OLED display;

The paragraph beginning on page 14, line 22 has been changed as follows:

Figures 6a to 6d show examples of well-filling according to embodiments of the ~~present~~ invention; and

On page 15, line 4 please insert a heading as follows:

#### DETAILED DESCRIPTION

The paragraphs beginning on page 15, line 5 have been changed as follows:

Referring now to ~~figure~~ Figure 6a, this shows a simplified vertical cross-section through a well 608 of a substrate 600 according to an embodiment of the present invention. The substrate includes an anode layer 606 on which are formed banks 610, faces 610a of the banks defining wall of well 608. As can be seen the faces 610a of banks 610 overhang the base of the well 608. In the substrate 600 of ~~figure~~ Figure 6a the bank angle is

approximately  $135^\circ$ , that is  $-45^\circ$  and height of a bank is approximately  $0.6\mu\text{m}$ . In figure Figure 6a the well is filled with a solution 602 of OLED material which, in this example, brims over the top of the well and forms a contact angle of approximately  $35^\circ$  with the top surface of a bank. Figure 6b shows the same substrate and well after the solvent has evaporated to leave a dry film 604 with the material together with small deposits on the tops of the banks adjacent the well-defining faces.

As can be seen from figures Figures 6a and 6b the capillary force around the pixel well edge pulls the ink 602 into the edges of the well, also giving good wetting into the well corners (not shown in figures Figures 6a and 6b) albeit with a slight ink overspill. Furthermore inaccurately placed droplets which land across the bank tend to be drawn into the well rather than drying on the bank. These effects are achieved with bank angles which are greater than the contact angle of the inkjet droplet. In practice this means angles of  $40^\circ$  or more, that is steep “positive” angles and “negative” angles. The degree to which fluid is pulled towards the edges of a well depends upon the angle of the bank, the viscosity of the fluid, and the contact angle of the fluid with the bank. A suitable angle can be determined by routine experiment, fabricating a range of wells with different bank face angles to see which results in the optimum performance. Generally it is desirable to obtain a substantially flat dry film 604 without too much material being drawn towards the edges of a well, thinning the film in the middle. The choice of a suitable bank face angle is described further below with reference to figure Figure 7.

Referring back to figure Figure 5b it can be seen that a drop 514 of dissolved material formed from a plurality of smaller droplets, once it grows to touch the sides of a well, will tend to be drawn into the corners. This allows molecular electronic material to be deposited



into a well by means of an incomplete filling method, that is where droplets are deposited so that they incompletely fill the well, and are then distributed to fill the well by capillary action.

To fabricate the undercut banks shown in ~~figures~~ Figures 6a and 6b a variety of techniques may be employed. Preferably a photodefinable polymer or photoresist such as polyimide or an acrylic photoresist is lithographically patterned using a mask or reticle and then developed to produce a desired bank face angle. Either a positive or a negative photoresist may be employed (for example there are image reversal methods which may be employed to reverse an image in a positive resist). To obtain an undercut photoresist the photoresist may be under-(or over-) exposed and overdeveloped; optionally an undercut profile may be assisted by soaking in a solvent prior to development. The skilled person will be aware that there are many variations of the basic spin, expose, bake, develop, and rinse procedure used in photolithography (see, for example, A. Reiser, *Photoreactive Polymers*, Wiley, New York, 1989, page 39, hereby incorporated by reference). Some particularly suitable resist materials are available from Zeon Corporation of Japan, who supply materials adapted for the fabrication of organic electroluminescent displays (negative resist materials in the ELX series, and positive resist materials in the WIX series).

The paragraph beginning on page 17, line 10 has been changed as follows:

It is has been observed that at the lower end of the preferred thickness range, with undercut banks, the edge of a bank tends to turn up slightly to form a lip, as shown in ~~figures~~ Figures 6c and 6d, which may improve containment of ink within a well. The formation of such a lip may be related to stress relief within the bank structure.

The paragraphs beginning on page 18, line 4 has been changed as follows:

This equation is helpful in understanding ~~figures~~ Figures 7b to 7e described below.

Figures 7b to 7e (not to scale) show effects of progressive increase bank face steepness; like elements of ~~figure~~ Figure 6 are indicated by like reference numerals. For each figure the left hand diagram illustrates a vertical cross section through a bank face forming the edge of a well containing dissolved molecular material 602. The centre diagram depicts the configuration of a drop straddling the bank edge, that is half on the bank face and half on the underlying anode.[[.]]

Referring first to ~~figure~~ Figure 7b, this shows a bank having an angle of approximately  $15^\circ$  to the underlying substrate, the liquid drop contacting the face of the bank at approximately  $35^\circ$ . Where a drop straddles the bank edge, one of the factors affecting the extent to which the drop is drawn into the well is the angle of the bank to the substrate. At shallow bank angles, the contact area between the bank face and the drop edge is relatively small. Consequently, there is a relatively small driving force for driving the drop from the low surface energy bank material and onto the higher surface energy well base.

The paragraphs beginning on page 20, line 6 have been changed as follows:

Thus the dry film thickness depends upon the height of the bank, the bank angle, the solvent evaporation (drying stage) conditions and the extent of any coffee-ring effect (also affected by the ink formulation, for example the solid content and molecular weight) and can be determined by experiment (for example by preparing films under a range of conditions thickness-distance graphs using an interferometer, for example from Zygo Corporation of Connecticut, USA. Referring to the centre diagram of ~~figure~~ Figure 7e it can also be seen

that there is a significant tendency for solvent carrying dissolved material to be drawn out from the sides of the drop along the undercut of the bank face, which is useful for obtaining substantially complete well-filling from incomplete or partial filling of a well by droplet deposition. Referring to equation 1 above and to ~~figure~~ Figure 7a, broadly speaking in the “ears” of the droplet  $\theta$  is reduced so that  $\cos \theta$  increases, effectively reducing the surface tension pulling the drop towards a more rounded shape.

The skilled person will ~~recognised~~ recognized that the above described techniques are not limited to use in the fabrication of organic light emitting diodes (small molecules or polymer) but may be employed in the fabrication of any type of molecular electronic device in which material is dissolved in a solvent and deposited by a droplet deposition technique. No doubt many effective alternatives will occur to the skilled person and it will be understood that the invention is not limited to the described embodiments encompasses modifications apparent to those skilled in the art lying within the scope of the claims appended hereto.